Supplement

Table S1. Characteristics of the campaign Periods (median and IQR)

periods parameters	period 1 (02. Feb 09:00 - 05. Feb 21:00)	period 2 (05. Feb 21:00 - 14. Feb 22:00)	period 3 (14. Feb 22:00 - 23. Feb 00:00)	short plume (18. Feb 17:00 - 19. Feb 07:00)
Dominant air mass origin	S to SW	NE to SE	W	NW
Local wind speed (m/s)	1.2 (0.8–1.7)	3.4 (2.7–4.1)	4 (2.5–6.3)	1.5 (1.2–1.7)
Gas ratio of SO_2 to NO_X	0.08 (0.04– 0.11)	0.63 (0.28– 0.83)	0.12 (0.09– 0.19)	0.09 (0.08– 0.09)
D_{modal_rBC} (nm)	190 (183–193)	239 (232–242)	181 (169–199)	242 (192–298)
Total aerosol concentration (μg m^{-3})	10.6 (8.7–11.7)	23.0 (20.4–27.2)	10.9 (8.1–15.8)	8.1 (6.6–10.2)
Mass fractions of organics (%)	36 (35–38)	37 (33–39)	29 (23–35)	28 (25–31)
Mass fractions of BC (%)	12 (10–16)	14 (12–15)	7 (4–11)	14 (9–21)
Mass fractions of nitrate (%)	24 (20–28)	18 (17–20)	35 (25–41)	27 (25–30)
Mass fractions of sulfate (%)	13 (11–17)	19 (17–22)	12 (11–15)	11 (10–12)

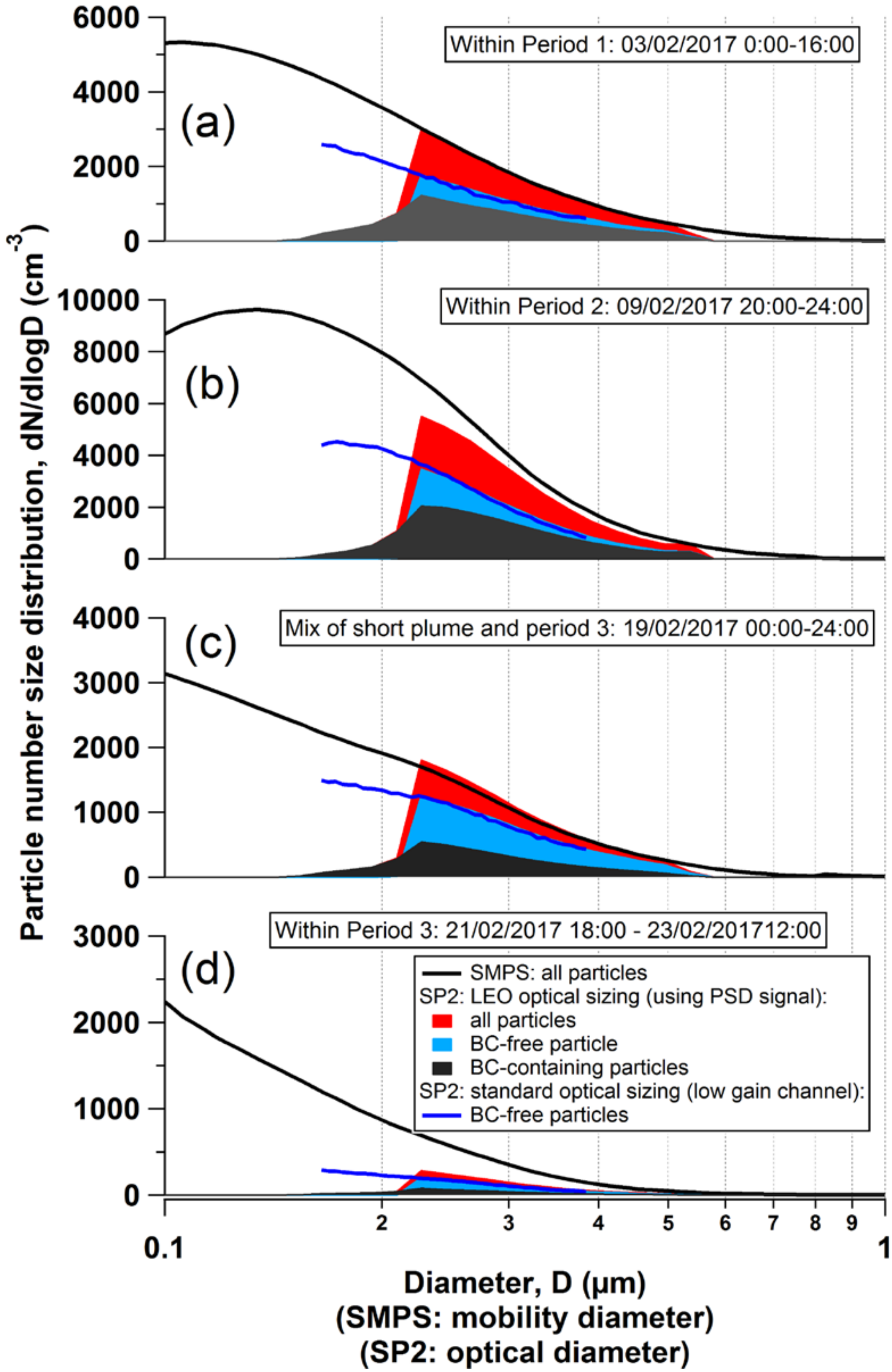

Within Period 1: 03/02/2017 0:00-16:00
(a)
Within Period 2: 09/02/2017 20:00-24:00
(b)
Mix of short plume and period 3: 19/02/2017 00:00-24:00
(c)
Within Period 3: 21/02/2017 18:00 - 23/02/201712:00
(d)
SMPS: all particles
SP2: LEO optical sizing (using PSD signal):
all particles
BC-free particle
BC-containing particles
SP2: standard optical sizing (low gain channel):
BC-free particles
Particle number size distribution, dN/dlogD (cm-3)
Diameter, D (μm)
(SMPS: mobility diameter)
(SP2: optical diameter)

Figure S1. Cross check of total particle number size distribution measured by SP2 compared with that from SMPS.

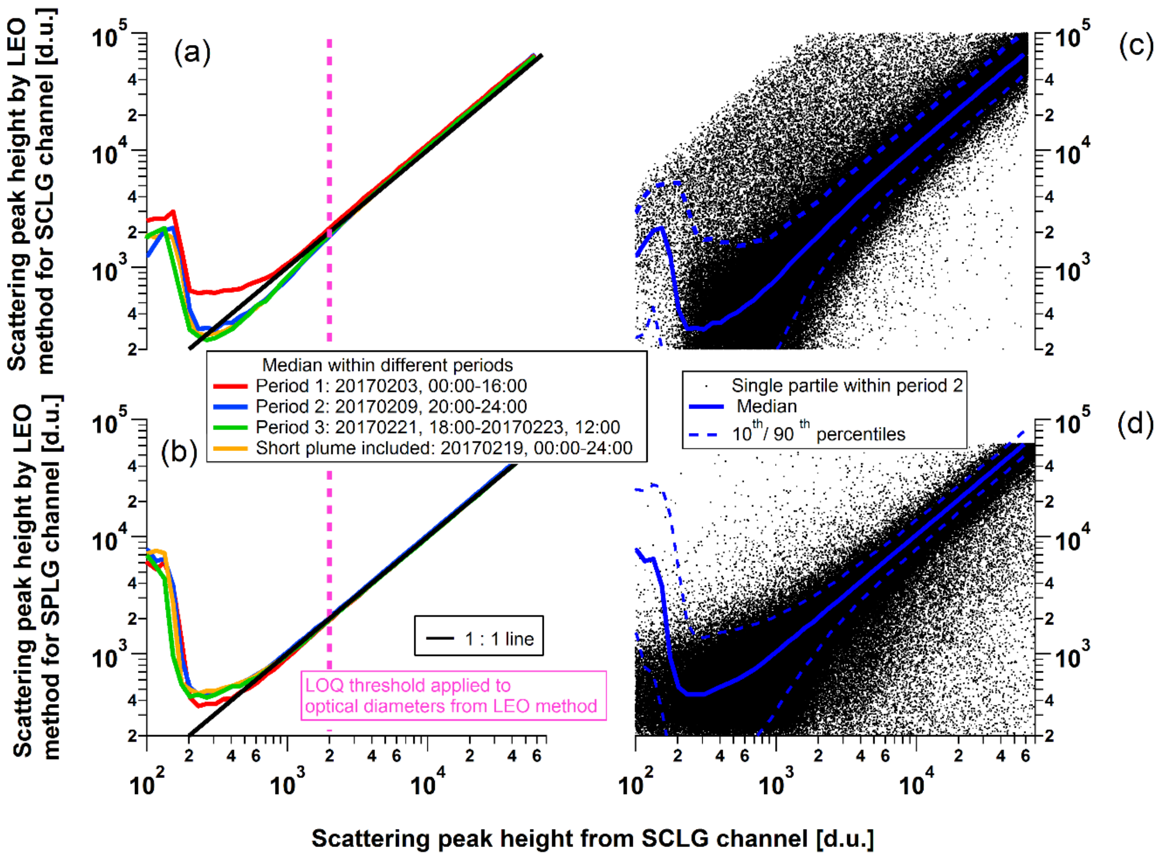


Figure S2. Verification of LEO method by BC-free particles. (a) Median of LEO scattering peak height vs standard scattering peak height from low gain scattering detector (SCLG) for four example days during different periods of the campaign. Scaling factors were applied to correct minor bias in the LEO fit analysis, i.e. to make the LEO fit results match the standard peak analysis. (b) LEO scattering peak height retrieved from the low gain position sensitive detector (SPLG) vs standard scattering peak height from the low gain scattering channel. (Adjustable scaling factors were applied to tie the LEO-fit to the calibration of the low gain scattering channel. (c) and (d) show single particle data corresponding to (a) and (b), respectively.

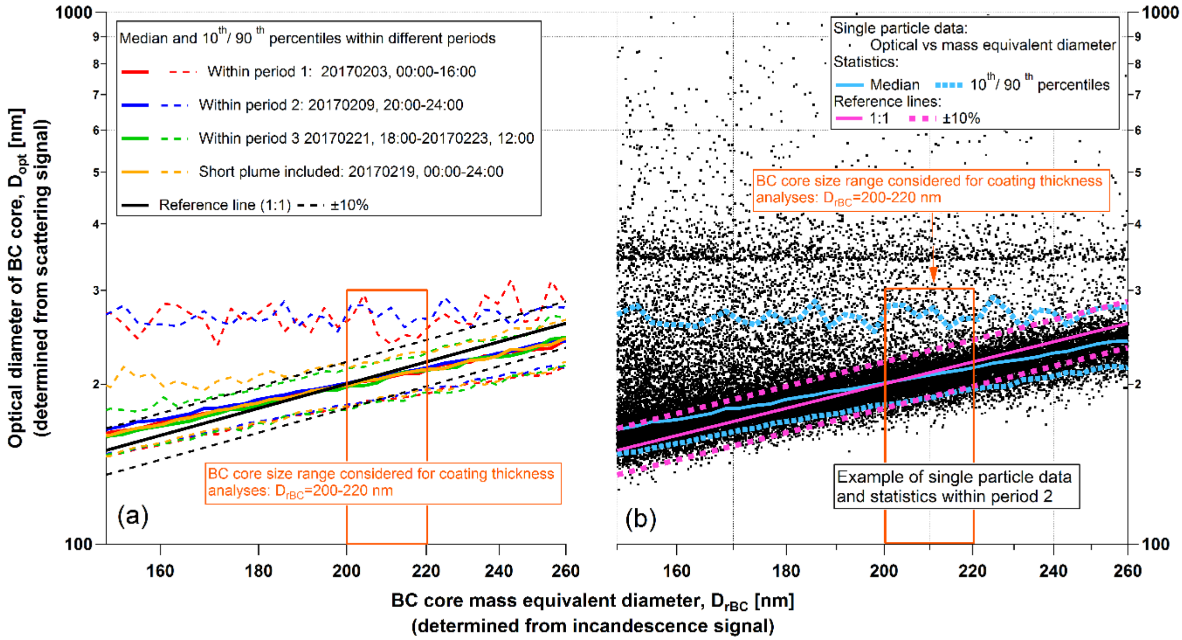


Figure S3. Verification of LEO fit: Optical diameter of the bare BC core compared with the rBC mass equivalent diameter. (a) Median and 10th and 90th percentiles of the single particle data. (b) Single particle data and corresponding statistics for period 2 as an example. The LEO fit results were used for the BC core diameter range from 200 nm to 220 nm, in which uncoated and coated particles can be sized optically. Within this size range the median values fall on the1 to 1 line within 2%. This ensures accuracy of the reported coating thickness for bare BC particles, i.e. that particle reported to have a coating thickness of zero indeed represent uncoated BC, except for the random noise present on single particle level.

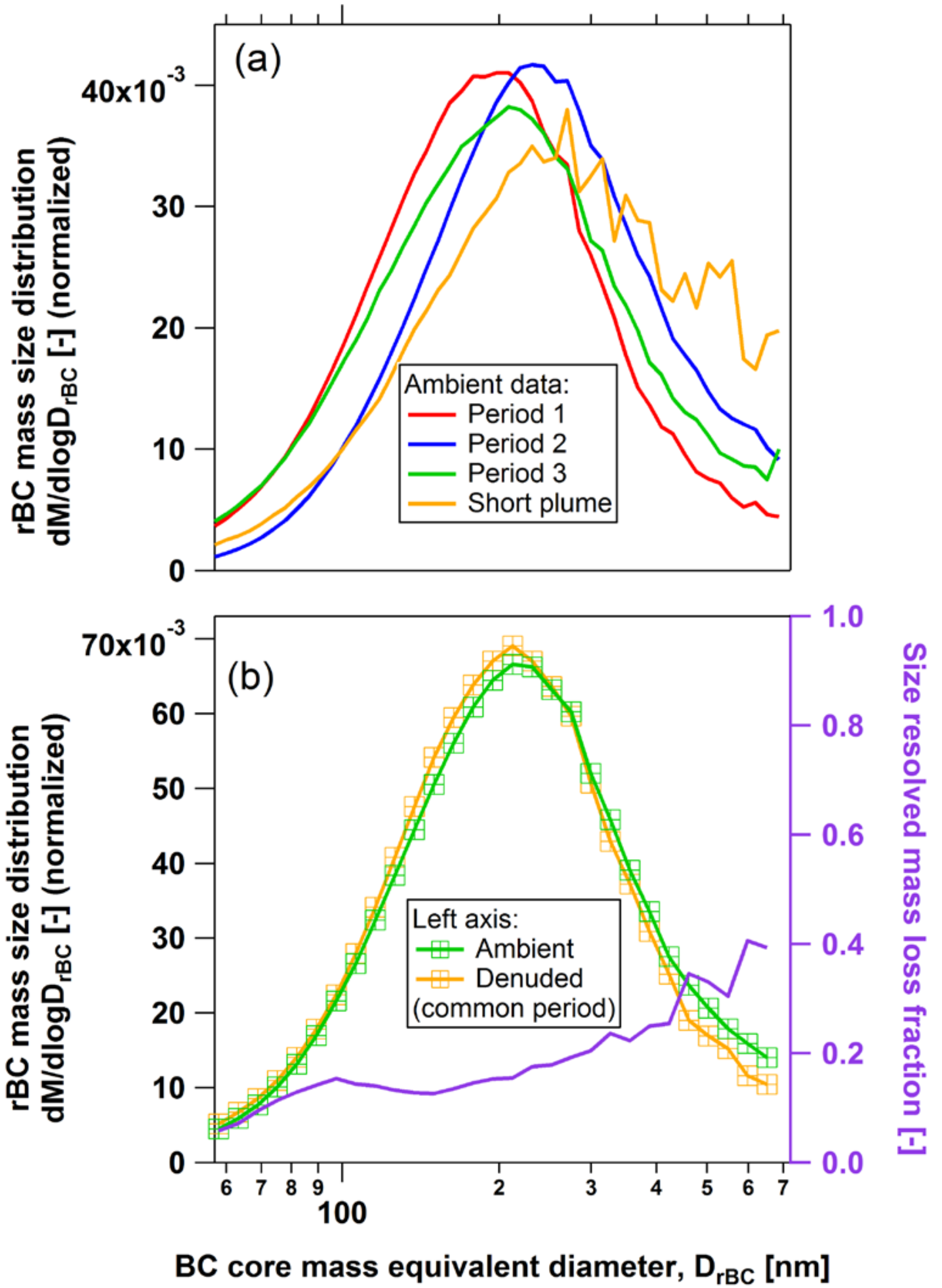


Figure S5. Normalized rBC mass size distributions as a function of BC core mass equivalent diameter. Ambient averaged for the different periods (a) and ambient compared with denuded data for complete common measurement period. The size resolved mass loss fraction within the catalytic stripper, computed from comparing denuded with ambient size distribution, is also shown in (b).

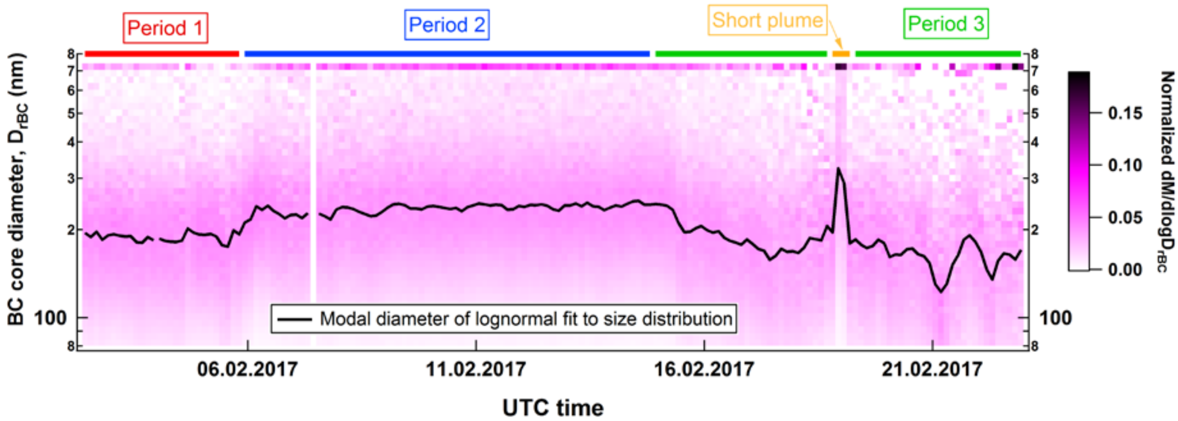


Figure S6. Time series of the normalized ambient rBC mass size distribution (at 3 h time resolution) as a function of BC core mass equivalent diameter (D_{rBC}). The overflow size bin including all BC particles D_{rBC} greater than ~700 nm is also shown.

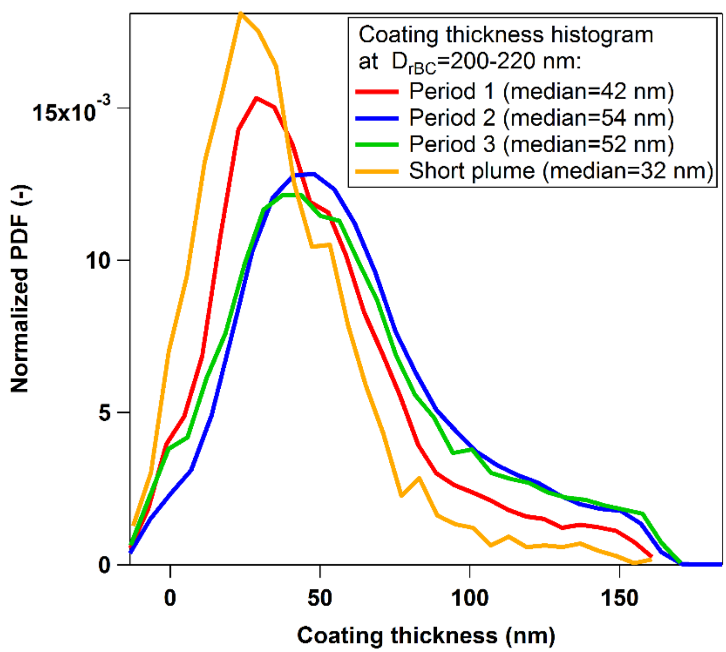
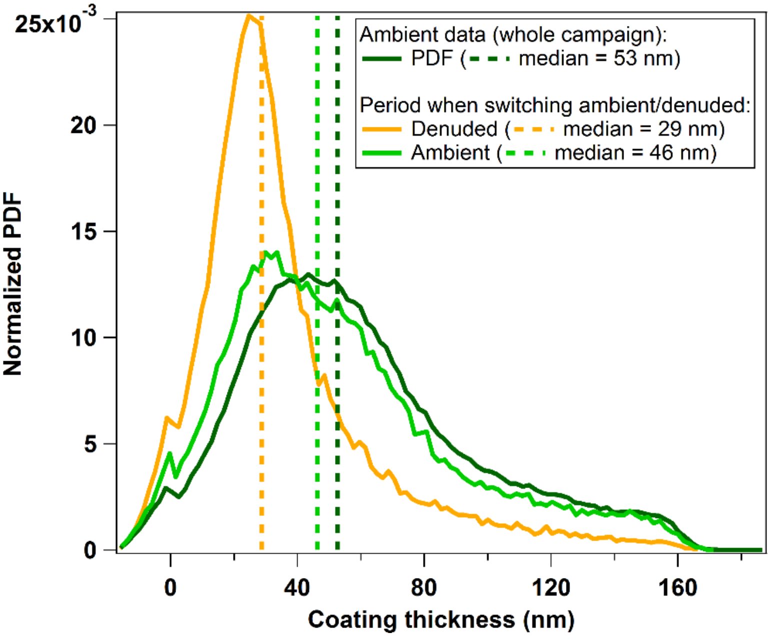


Figure S7. Histograms of BC particle coating thickness on single particle level separately shown for the three periods and the short plume.



985 **Figure S8. Histograms of BC particle coating thickness on single particle level for rBC mass equivalent core diameters between 200 nm to 220 nm. Histograms are separately shown for the whole campaign (ambient sample only) and for the period when the denuder was operated (ambient and denuded samples).**

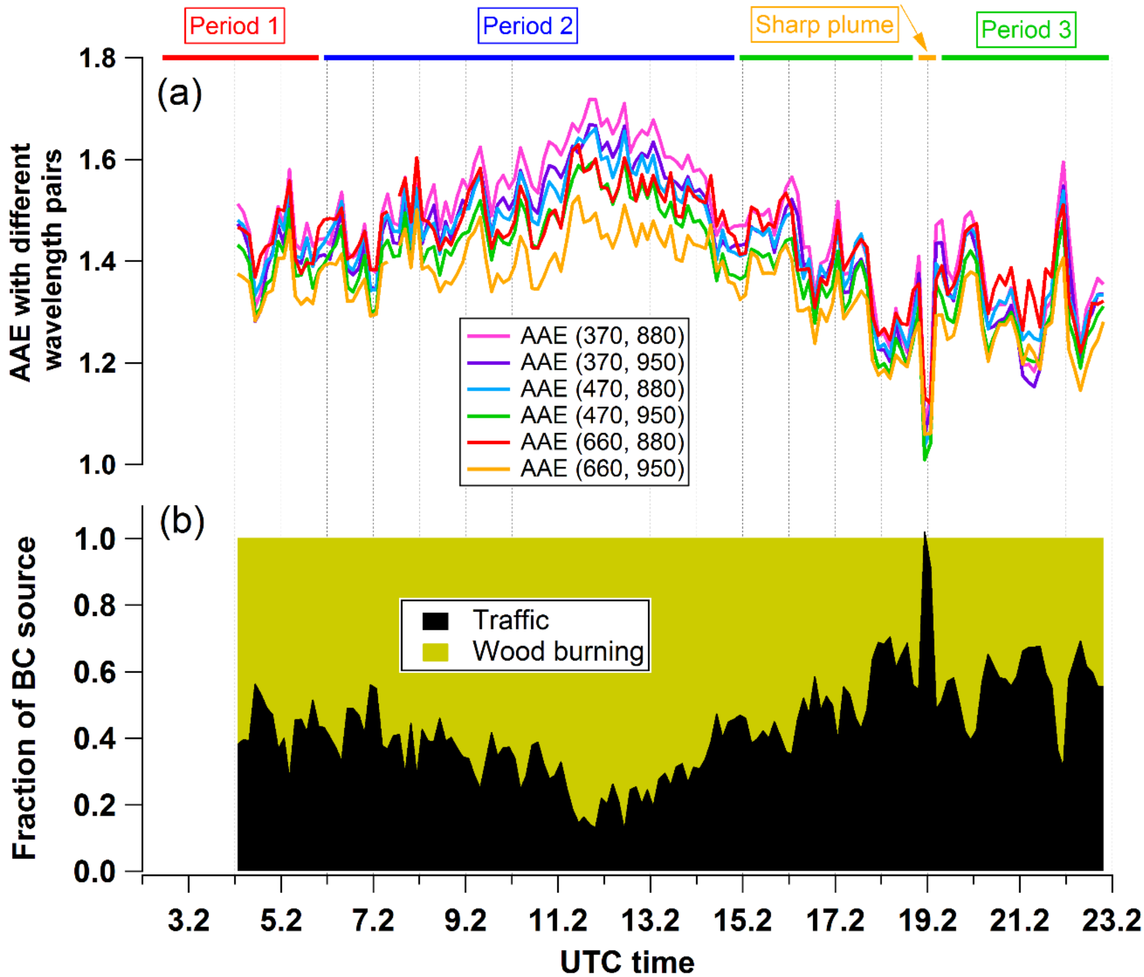


Figure S9. (a) Time series at 3 h time resolution of AAE of different pairs of wavelengths, and (b) the estimated fractional contribution of traffic and wood burning emissions to BC mass. The attribution of sources was done using the so-called "aethalometer model" using the coefficients reported in Zotter et al. (2017). Note that traffic and wood burning fractions are upper limits as BC from coal burning, which is potentially present, could be assigned to either source.